Mineralogy, Petrology, and Geochemistry of the Lunar Samples

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On July 24, 1969, the first extraterrestrial samples, with the exception of meteorites, were returned to earth by Apollo 11. Since then these samples and the samples returned by Apollo 12 have been subjected to scientific investigations by hundreds of scientists from many countries. Drawing on advances from the last 25 years of study of meteorites and terrestrial rocks the variety and sophistication of the techniques used on these samples is truly impressive. It can truthfully be said that 10 years ago we could not have made the measurements, and that, even if we had had the data, we could not have interpreted it. The electron microprobe, the scanning electron microscope, and the mass spectrometer, as well as other instruments and techniques, have joined the microscope as routine tools with which to attack a petrologic problem.

Most of the results to date have been published in special issues or volumes that are referenced separately in the bibliography (with tables of contents). Basic data on the flights, on the sample collections, and on the experiments conducted on the lunar surface are contained in two National Aeronautics and Space Administration reports (Ref. B and Ref. F; see also Ref. A and Ref. E). The papers presented at the Apollo 11 Lunar Science Conference in January, 1970 are contained in a special issue of *Science* (Ref. C). Expanded and revised versions of most of these papers are contained in a three-volume, 2492-page supplement to *Geochimica et Cosmochimica Acta* (Ref. D). Fourteen papers describing Lunar Sample 12013 have been collected in a special issue of *Earth and Planetary Science Letters* (Ref. G). (The abbreviations Ref. A, . . . , Ref. G replace the date in references to papers contained in these special compilations.)

Most of the basic observations, data, and conclusions summarized here were obtained by dozens of investigators. Hence, the references are included mainly to indicate a good starting point for further reading and are not inclusive.

The astronauts on the Apollo 11 and 12 missions did not sample bed rock. The returned lunar samples included large fragments of igneous rock and microbreccia collected on the lunar surface and samples of lunar soil.

The lunar soil is a diverse mixture of crystalline and glass fragments with a wide range of size. Most of the lithic fragments in the soil are similar to the larger igneous rock samples and are probably derived from the underlying bed rock. However, some are totally different from the larger fragments and may represent samples from other parts of the moon. The abundance of anorthosite fragments has led a number of workers to infer that the lunar highlands are comprised of anorthosite and that the separation of plagioclase from the magma to form the highlands is a major lunar process [*Smith, et al.*, Ref. D, 937–956; *Smith, et al.*, 1970; *Wood, et al.*, Ref. D, 965–988].

The glass fragments include regular forms, such as spheres and dumbbells, as well as broken fragments and irregular forms. Irregular splotches of glass represent splashes of molten silicate. Surfaces of both rock and glass fragments show beautifully preserved pits as small as a few microns in diameter, which are the result of impacts by tiny hypervelocity particles. Small fragments of meteorites have been found in the soil, but many of the pits may have formed in the hypervelocity stream of particles and melt droplets thrown out from larger impact features. The glass fragments shown a broad range in color and in chemical composition, which exceeds that of the larger igneous rocks. Broad studies of the soil materials include those of *Carter and MacGregor* [Ref. D, 247–266], *Duke et al.*, [Ref. D, 347–362], *Frondel et al.* [Ref. D, 445–474], *Wood et al.* [Ref. D, 965–988], and *Wood et al.* [1970].

The microbreccias are a mechanical mixture of rock and glass fragments, not unlike the soil, but are compacted or welded into a coherent rock. Like the soil, they also contain rock types not found among the larger igneous rock samples. A much smaller proportion of microbreccia samples was returned from the Apollo 12 site than from the Apollo 11 site.

The larger igneous rock fragments from the Apollo 11 site range from very fine-grained vesicular basalts to vuggy, medium-grained microgabbros with little difference in mineralogy. A summary description of the rock samples is given by *Schmitt et al.* [Ref. D, 1–54]. These rocks consist predominantly of Ti-rich augite, calcic plagioclase, and ilmenite with lesser amounts of olivine, cristobalite, troilite, Fe-metal, and an interstitial K- and Al-rich 'phase,' consisting predominantly of glass and fine-grained crystalline material [*Roedder and Weiblen*, Ref. D, 801–837]. The augite has a large range in composition, ranging in single crystals from about $(Ca_{35}Fe_{15}Mg_{50})SiO_3$ to a $(Ca_{13}Fe_{85}Mg_2)SiO_3$ composition with a pyroxenoid structure. The augite also contains 2–10% of $CaTi(Al_2O_6)$ end member and in some rocks the compositional range is further extended by exsolution of pigeonite. Plagioclase grains are fairly homogeneous with compositions ranging from about An_{75} to An_{98}. A good introduction to the dozens of detailed papers on the petrology and mineralogy of the lunar igneous rocks is provided by *Agrell et al.* [Ref. D, 81–86], *Brown et al.* [Ref. D, 195–220], *Keil et al.* [Ref. D, 561–598], *Smith et al.* [Ref. D, 897–926], *Weill et al.* [Ref. D, 937–956], or *James and Jackson* [1970].

Most of the igneous rocks returned from the Apollo 12 site are generally similar, but they exhibit a wider

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range of modal composition, grain size, and texture. Low-Ca pyroxenes and olivine are more abundant. Olivine vitrophyre contains skeletal olivine, which may represent subsolidus crystallization.

Minor minerals include zircon, baddeleyite, chlorofluoroapatite, whitlockite(?) high in rare earth elements, tridymite, quartz, K-feldspar, various spinels, metallic copper, pseudobrookite, and dysanalyte (?). Armalcolite (a ferropseudobrookite), pyroxferroite (an iron-analog of pyroxmangite), and several spinels of unusual composition are new minerals unknown in terrestrial rocks [*Anderson et al.*, Ref. D, 55–64; *Chao et al.*, Ref. D, 65-80; *Agrell et al.*, Ref. D, 81–86].

Melting experiments on silicate melts with chemical compositions similar to the Apollo 11 rocks indicate that the major mineral phases crystallized from an igneous melt in a narrow (~ 75°C) temperature interval within the temperature range 1210° to 1075°C; minor interstitial liquids continued to crystallize down to at least 1045°C [*Smith et al.*, 1970; *O'Hara et al.*, Ref. D, 695–710; *Roedder and Weiblen*, Ref. D, 801–837; *Ringwood and Essene*, Ref. D, 769–800]. Microscopic and microprobe examination clearly demonstrates that an interstitial residual liquid persisted, which was rich in Si, K, Al, and various minor elements and which was probably immiscible with the main melt [*Roedder and Weiblen*, Ref. D, 801–837]. An immiscible sulfide liquid, from which the troilite and metallic iron crystallized, probably separated during the later stages of crystallization [*Skinner*, Ref. D, 894].

Viscosities are much lower and densities are much higher for lunar magmas than for typical terrestrial basaltic magmas [*Weill et al.*, Ref. D, 937–956; *Murase and McBirney*, 1970]. These differences are significant to an understanding of the textural feature and magmatic differentiation in the lunar rocks and of the morphological features of the lunar surface. For example, owing to low viscosity and high density, an individual lava flow may form a very thin cover over a considerable portion of a mare surface.

Experiments indicate that the observed phases could have coexisted only in a very dry, highly reducing system. The partial pressure of oxygen is estimated as 10^{-16} atm [*Smith et al.*, Ref. D, 917), 5 orders of magnitude lower than that for typical terrestrial basaltic magmas. The low level of ferric iron in pyroxenes, determined by Mössbauer spectroscopy and electron spin resonance, the presence of troilite and native iron, and the occurrence of the unusual spinel compositions are further evidence of the low oxidation state of the magmas. The silicate minerals are highly transparent and clear owing to the complete lack of any hydrous alteration or weathering. No minerals have been shown to contain hydroxyl. D/H measurements indicate that the bulk of the H is of solar-wind origin and that it is most likely that the lunar surface is nearly devoid of primary water [*Epstein and Taylor*, Ref. D, 1091]. Vesicles and vugs are common features in the lunar rocks, but the composition of the volatile phase is not known. Despite the abundance of troilite, calculations indicate that vesiculation due primarily to S_2 pressure is unlikely [*Weill et al.*, Ref. D, 949–950].

Experiments at high pressures and temperatures show that the Apollo 11 rocks would have densities at relatively shallow depths that fa- exceed the density of the moon [*Ringwood and Essene*, Ref. D, 769–799]. Hence, as predicted by *Wetherill* [1968], rocks with the composition of the Apollo 11 rocks cannot represent the bulk composition of the moon and must represent a derivative of some more primative material.

The chemical composition of the Apollo 11 igneous rocks is not grossly unlike that of terrestrial basalts, but the Fe and Ti content is distinctly higher and the Na content distinctly lower than occurs in typical terrestrial basalts. The compositions are quite uniform, but differ in the content of certain trace elements not abundant in the major minerals. The content of these 'incompatible' trace elements divides the rocks into two distinct groups with K differing by a factor of 4.4, Rb by 7.7, Ba by 3.0, Ce by 2.1, Zr by 1.5, Hf by 1.6, U by 4.1, and P by 2.0 [*Gast et al.*, Ref. D, 1151]. Each of these groups is so similar chemically and isotopically that it appears likely that only two igneous bodies were sampled on the Apollo 11 mission. Compared with terrestrial basalts, they are all richer in refractory elements such as Ba, Y, Ti, Hf, Ta, Th, U, and trivalent rare earth elements, lower in alkalies, lower in volatile and chalcophile elements such as Bi, Hg, Zn, Cd, Tl, Pb, Ge, and Br, and lower in most siderophile elements including Co, Ir, Ni, Pd, and Au [*Ganapathy et al.*, Ref. D, 1133]. The pattern of the rare earth elements is similar to that in chondritic meteorites, but is distinguished by a marked depletion of Eu relative to other rare earths. This has been attributed to extensive separation of plagioclase [*Philpotts and Schnetzler*, Ref. D, 1471–1486].

The chemical compositions of rocks from the Apollo 12 site differ in detail, but show these same characteristics and correspond most closely to the low-K group.

The chemical composition of the soils and breccias contains many of the distinctive characteristics of the igneous rocks, but elemental and isotopic abundances clearly indicate that it contains at least two other components. Enrichment in Ni and in elements characteristic of carbonaceous chondrites, such as Cd, Zn, Ag, Au, Cu, and Tl, indicate a maximum meteoritic contribution of 1–2% [*Ganapathy et al.*, Ref. D, 1117–1142]. Another component is high in 'incompatible' elements [*Goles et al.*, Ref. D, 1177–1194] and corresponds closely to the difference in composition between the two Apollo 11 rock groups, to the composition of the interstitial, residual-liquid phase of the basalts, and to several exotic fragments that have been described, notably rock 12013 (see Ref. G) and Luny Rock #1 [*Albee et al.*, Ref. C, 463–465, *Albee and Chodos*, Ref. D, 135–158].

Model Sr/Rb ages for the soil are much older than the ages of the rock fragments contained within it and indicate the presence of a component with a high Rb/Sr ratio with a model age of 4.6×10^9 years. Similarly model Pb-U-Th ages also indicate the presence of an ancient component with high Pb-U-Th contents.

Rock 12013 (described in Ref. G) is important

because it has many of the characteristics of these soil components and it provides clear evidence of the existence and importance of K-, Rb-, U-, Th-rich granitic rocks and magmatic reservoirs in the early history of the moon. Rock 12013 contains the highest Si content and 10–50 times more K, Rb, Li, Ba, Y, Zr, Th, U, and rare earth elements than other lunar rocks. It is an extremely heterogeneous fragmental rock that has been permeated by a once-fluid granitic component, consisting dominantly of potassic feldspar and quartz. This granitic component has a high Rb/Sr and a model age of about 4.5×10^9 years. The existence of this rock and similar exotic fragments, taken in conjunction with the soil composition, indicates that such rocks will be found elsewhere on the moon and that they are important to our understanding of the early differentiation history of the moon.

Rock 12013 is also important in finally ruling out a lunar origin for tektites [*Taylor and Epstein*, Ref. G, 208–210]. Although it has an SiO_2 content that is in the same range as that of some tektites, the oxygen isotopic values are totally outside the range of values for tektites.

No simple process can produce a melt with the distinctive characteristics of the lunar rocks from any known terrestrial rock or meteorite compositions. Volatile and alkali element loss in heating, gross separation of metal and sulfide phases, and silicate melt-solid differentiation must all be important [*Ringwood and Essene*, Ref. D, 769–799; *Ganapathy et al.*, Ref. D, 1117–1142].

The Rb/Sr isotopic data on the soil and on these exotic fragments strongly suggest that the distinctive chemical patterns were present almost at the beginning of the moon's history [*Lunatic Asylum*, Ref. G, 137–163], and, taken together with other geochemical data, appear to rule out a fission origin. Models involving accretion in a planet-satellite system, with the accretion dependent on temperature, the mass of the bodies, or both, have been proposed to account for many of these characteristics [*Ringwood and Essene*, Ref. D, 769–800; *Ganapathy et al.*, Ref. D, 1117–1142].

However, the isotopic, chemical, and petrological data require, in addition, rapid differentiation of a lunar crust at 4.5×10^9 years and a mechanism for melt formation at 3.3×10^9, 3.6×10^9, and possibly 4.0×10^9 years. It is not yet clear whether the necessary heat source is internal, from long-lived radioactivities, or external, by very large scale impact. Nor is it yet clear whether the entire moon has been differentiated or only its outer shell.

A large number of important studies, which do not fall directly into petrologic, mineralogic, and geochemical categories, were performed on the lunar samples and are included in References C and D. The studies of physical properties are important to the interpretation and extension of telescopic observations, seismic experiments, and magnetic measurements. Rocks on the lunar surface serve as a monitor of radiation from both the galaxy and the sun, and detailed studies have provided new information on lunar surface processes, the cosmic-ray flux, and the solar wind. The intensive search for complex carbon compounds of protobiological significance and for viable organisms produced negative results, and most of the observed carbon appears to be of solar-wind origin.

Although collected from only two locations, the Apollo samples have given a vast new insight into the processes that formed the moon and shaped its surface. The results do not resolve the problem of the origin of the moon. However, significant limits have been established for the chemical parameters, on the time, and on the rate of many processes. A large number of constraints must now be met by any theory that attempts to explain the origin of the moon.

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Radioactive and Radiogenic Isotope Research

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The number of laboratories and workers in the field of radioactive isotope research has grown rapidly over the past four years. With this growth has come a large increase in the volume of literature on the subject. Consequently the bibliography given below is meant to include the more significant or more complete investigations. The references are restricted to papers published by U.S. laboratories from approximately January 1967 through October 1970.

Extraterrestrial Studies. The availability of returned lunar material has stimulated many kinds of investigations. The great antiquity of lunar materials has been one of the most interesting findings from the first studies. The Rb-Sr age of crystalline rocks from the Apollo 11 site on the Sea of Tranquillity is about 3.7 billion years [*Papanastassiou et al.,* 1970; *Gopalan et al.,* 1970; *Hurley and Pinson,* 1970], a value in close agreement with the highest K-Ar ages found by *Funkhouser et al.* [1970] and by G. Turner at Sheffield in some excellent $^{40}Ar/^{39}Ar$ dating. The isotopic lead ages [*Tatsumoto,* 1970*a, b*; *Silver,* 1970] are more difficult to interpret. The problem is partly that all leads found on the moon to date are highly radiogenic, making it difficult to know the proper initial lead correlation. Also the amount of common lead found in the rocks is so small that laboratory contamination may be critical to some interpretations. The lead ages are nearly concordant at 4.1 b.y. if initial lead corrections are based on primordial or modern terrestrial lead; however, Tatsumoto has shown how the data can be reconciled with ages of 3.4 to 3.8 b.y. on the basis of two-stage evolution models for the lead. Such models can remove the conflict with the Rb-Sr and K-Ar data.

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The data from the Apollo 11 dust and breccia yield a formal Rb-Sr isochron age of 4.6 b.y. [*Papanastassiou et al.,* 1970], whereas the isotopic lead ages are nearly concordant at 4.7 b.y. [*Tatsumoto,* 1970*b*; *Silver,* 1970; *Gopalan et al.,* 1970]. In spite of the close agreement of the two ages and their agreement with Rb-Sr ages from meteorites (see below), it is not yet possible to state that the soil data record the age of the moon.

Few Apollo 12 data (Ocean of Storms) have appeared as yet. The Rb-Sr age of the unusually high potassium rock 12013 is 4.0 b.y. [*Lunatic Asylum,* 1970; *Schnetzler et al.,* 1970], whereas the isotopic lead age is about 3.9–4.0 b.y., according to a two-stage lead evolution model [*Tatsumoto,* 1970*a*]. Turner reports $^{40}Ar/^{39}Ar$ ages of 3.9 b.y. for both light and dark phases of the rock.

D. A. Papanastassiou and G. J. Wasserburg (preprint) report Rb-Sr ages of 3.3 b.y. for two crystalline rocks at the Apollo 12 site, and a Rb-Sr model age of 4.44 b.y. for dust. The younger crystalline rock ages at the Apollo 12 site, compared with the Apollo 11 site, may be reflected in the dust ages at the two sites, but the most impressive aspect of the ages is their general agreement.

Several important advances in meteorite studies merit special mention. Rb-Sr isochrons have been determined for each of the various chondrite groups, H, L, LL, E, and C [*Gopalan and Wetherill,* 1968, 1969, 1970; *Kaushal and Wetherill,* 1969, 1970]. Their work relied almost entirely on falls to eliminate effects due to possible chemical alterations of finds. Ages varied from 4.48 to 4.69 b.y., analytical errors in each case being about ±0.15 b.y. The results are thus consistent with Rb-Sr differentiation ages of 4.55 and 4.60 b.y. for all the groups. *Bogard et al.* [1967] obtained a Rb-Sr age of 4.7 ± 0.1 b.y. for the Norton County achondrite, using separated fractions from the stone. This is one of very few stones for which satisfactory internal isochrons are measured. New data on the Rb-Sr ages of silicate inclusions are given by *Burnett and Wasserburg* [1967*a, b*]. Six irons yield ages of around 4.5 b.y., a value similar to the Rb-Sr ages of stone meteorites but not in agreement with the older K-Ar ages that have been reported for the metal phase of iron meteorites. *Rancitelli and Fisher* [1968] believe that the older K-Ar ages result from potassium migration. Silicate inclusions from the Kodiakanal iron meteorite give a Rb-Sr age of 3.8 ± 0.1